

PATENT SPECIFICATION

NO DRAWINGS

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COMPLETE SPECIFICATION

Improvements relating to Iso-Indolone Dyes and their use

We, J. R. GEIGY A.-G., a body corporate organised according to the laws of Switzerland, of 215 Schwarzwaldallee, Basle, Switzerland, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The invention concerns new coloured condensation products which are distinguished by their colour strength, their stability to heat, their fastness to light and the fastness to migration of the coloured plastics obtained therewith. The invention also concerns a new production process which, by using new starting materials, enables new coloured condensation products to be produced in greater purity and good yields. The invention concerns, in addition, processes for the dyeing of organic material such as paper and other types of cellulose, in particular however, processes for pigmenting plastics and lacquers by the use of the new condensation products as well as the material dyed with the aid of these products.

It is known that by condensing 3-imino-isindole-1-one, 3-thio-isindole-1-one or 3-imino-isindole-1-thione and derivatives thereof substituted at the benzo radical with aromatic primary amines and hydrazines which, apart from the amino group or the hydrazino group, contain also a further primary amino or hydrazino group or a substituted amino or hydrazino group, coloured condensation products are obtained which are suitable for the dyeing of cellulosic material such as paper, for the dyeing of lacquers as well as for pigmenting plastics. In the known process, the substituent in the 3-position of the isindole is split off in the form of ammonia or hydrogen sulphide on heating the reactants alone or in inert organic solvents or diluents such as glacial acetic acid or alcohols and a C=N— double

linkage is formed between the amino nitrogen atom of the aromatic amine and the carbon atom in the 3-position of the isindole ring. However, condensation products which contain more than two substituents in the benzo radical of the isindolone have not been known according to this process.

According to the present invention, however, it has now been found that the condensation products obtained from polyprimary amino compounds of the homo- or hetero-cyclic aromatic series or hydrazine and isindole-1-ones which contain at least three halogen atoms in the benzo radical, in particular three and, advantageously four halogen substituents, are distinguished in the pure state from similar known compounds by particularly favourable fastness properties. For example, their colour is stronger, they are more fast to migration in plastics and, in particular, more fast to light. They are obtained by condensing one mol of polyprimary amino compounds of the homo- or hetero-cyclic aromatic series or hydrazine with a molecular equivalent for every primary amino group thereof of the same or different isindole-1-ones which contain at least three halogen atoms in the benzo radical, advantageously tetrahalogenated, which isindole-1-ones contain in the 3-position an imino group which can be organically substituted, a doubly bound sulphur atom, two halogen atoms, two secondary amino groups or two other groups.

The isindolone derivatives used according to the present invention contain, according to the definition, at least three halogen atoms in the benzene ring, advantageously such as chlorine, bromine or fluorine. Of technical importance are the tetrahalogen and of these, particularly the tetrachloro compounds. These isindolone derivatives which contain at least three halogen atoms in the benzo radical are new. The 3-imino compounds are obtained,

[Price]

e.g. from tri- or tetra-halogen phthalic acid anhydride on heating in nitrobenzene with urea and ammonium molybdate. However, the purity of 3-imino-isindole-1-ones so produced leaves something to be desired. Isoindolone compounds which are more pure and therefore more suitable for reaction in the process according to the present invention are obtained by a new method from phthalimides which contain at least three halogen atoms in the benzene ring by reacting with at least 1 mol of phosphorus pentachloride in inert organic solvents at a raised temperature while excluding moisture. 3,3-Dichloro-isindole-1-ones are obtained which can be used either direct or which can first be converted with ammonia or primary organic amines into the also very reactive, corresponding 3-imino compounds. Also the known 1,3,5-trichloro-isindolenines which contain at least three halogen atoms in the benzo radical can be used as starting materials. These compounds are obtained from the corresponding phthalimides by reacting with 2 mols of phosphorus pentachloride in the warm while excluding moisture and hydrolysed before or during the reaction with amines, advantageously by using water-containing organic solvents, to form the corresponding 3,3-dichloro-isindole-1-ones. The hydrolysis is performed carefully in the cold or at slightly raised temperature. It is most practical to react 1 mol of tri- or tetra-chlorophthalimide with 1.1 to 1.3 mol of phosphorus pentachloride and to use the 3,3-dichloro-isindole-1-one compound which crystallises out on cooling and is filtered off. However, the reaction solution can also be used direct if, before or during the condensation with the amino compounds, care is taken to ensure the hydrolysis of the 1,3,5-trichloro-isindolenines which are also formed by conducting the condensation in the presence of moisture.

The following polyprimary amino compounds can be used for example in the production of coloured condensation products according to the present invention: hydrazine and advantageously aromatic polyprimary amino compounds of the homo- or heterocyclic series, e.g. 1,3- and 1,4-diaminobenzenes, 4,4'-diaminodiphenyl- and -triphenyl alkanes, 4,4'-4''-triaminotriphenyl methane, 4,4'-diaminodiphenylamine, 4,4'-diaminodiphenyl, 4,4''-diaminotriphenyl, 2,4,4'-triaminodiphenyl, 4,4'-diaminostilbene, 1,4-, 1,5-, 1,7- and 2,6-diaminonaphthalenes, 2,7-diaminofluorene, 1,4- and 1,5-diamino anthraquinones, 2,6-diamino pyridines, 2,7-diamino-carbazole, 2-(4'-amino-phenyl)-6-amino-benzthiazole, 2,6-diaminobenzthiazole, 2,5-bis-(4'-amino-phenyl)-oxadiazole and thiadiazole. Naturally, also homo- and heterocyclic aromatic polyamines in which the amino groups are in other positions can be used. Advantageously, however, diamino compounds are used in which the amino groups are in the position of possible

quinones. All aromatic rings can be still further substituted, e.g. by alkyl groups such as methyl, ethyl, tert. butyl, tert. amyl groups, by ether groups such as methoxy, ethoxy, butoxy, phenoxy, methylmercapto, ethylmercapto, phenylmercapto groups, by halogen atoms, by sulphonic acid and carboxylic acid groups, their salts, esters and amides, by nitro, acylamino, secondary and tertiary amino groups. Particularly favourable for the colour strength of the condensation products according to the present invention is the choice of such diprimary amino compounds which, on joining the isindolone rings, produce a system of conjugated double linkages. Examples of such diamino compounds are hydrazine, 1,4-diaminobenzene, 1,4'-diaminodiphenyl and 4,4'-diaminostilbene compounds as well as diamino compounds of more highly condensed homocyclic or heterocyclic structures in which the amino groups take the positions of possible quinones. The list given above contains some examples of such diamino compounds. Of technical interest are the 1,3- and 1,4-diaminobenzene, 4,4'-diaminodiphenyl, 4,4'-diaminostilbene derivatives and the diamionaphthylene derivatives corresponding to quinones, also the derivatives of 4,4'-diaminodiphenyl compounds which, by ring-forming linking members, are further condensed to form fluorenes, diphenylene oxides, diphenylene sulphides, diphenylene sulphones, and carbazoles. All these compounds may carry further substituents in the aromatic rings.

The condensation of these polyprimary amino compounds with the reactive isindole-1-ones substituted in the 3-position proceeds very quickly and easily often even in the cold, or on warming the well mixed components, particularly advantageously in the presence of inert organic solvents and diluents such as hydrocarbons, halogenated hydrocarbons, aliphatic or alicyclic ethers and ketones or esters. For example cyclohexane, benzene, toluene, xylenes, tetrahydronaphthalene, naphthalene, diphenyl, carbon tetrachloride, tetrachloroethylene, chlorobenzene, di- and tri-chlorobenzene, nitrobenzene, dibutyl ether, dioxan, diphenyl ether, acetone, cyclohexanone can be used as inert solvents. It is possible to perform the condensation in the presence of acid-binding agents such as sodium acetate, magnesium oxide or calcium carbonate. Also organic nitrogen bases may be used, e.g. pyridine, quinoline or dialkyl anilines.

When they contain no acid, salt-forming groups, the coloured condensation products according to the present invention are very difficultly soluble substances. They precipitate from the reaction solution even while hot and can easily be purified by filtering off and washing with organic solvents, water, aqueous alkalis and aqueous acids. If starting materials are used which contain acid salt-forming groups, e.g. sulphonic acid or carboxyl groups,

the condensation is performed advantageously in solution in a mixture of an organic solvent with water. Yellow, orange, red, brown or violet to black condensation products are obtained according to the polyamide used. When they are insoluble in water, due to their excellent fastness to light, their colour strength and fastness to migration in plastics, such as polyvinyl chloride, they are valuable pigment dyestuff which are excellently suited for pigmenting lacquers, polymeric synthetic materials and rubber. In finely dispersed form, they can also be used for the dyeing of paper or for the dyeing of viscose or cellulose esters or polyamides in the spinning mass. If they contain water solubilising, salt-forming groups, they can be converted into insoluble dye lacquers with the calcium, barium, strontium, aluminium or heavy metal salts usual for this purpose or, depending on the affinity to cellulose or polypeptide fibres, they can be used for the dyeing of cellulosic material such as paper, cellulose fibres or animal fibres.

The following Examples illustrate the invention. Where not otherwise stated, parts are given as parts by weight. The temperatures are in degrees Centigrade.

EXAMPLE 1

A solution of 5.4 parts of 1,4-diaminobenzene in 200 parts of *o*-dichlorobenzene is added to a solution of 34 parts of 3,3,4,5,6,7-hexachloro-isoindole-1-one in 300 parts of *o*-dichlorobenzene. The yellow suspension which immediately forms is then heated for 3 hours at 160–170° whereupon it is filtered hot and the residue is washed with *o*-dichlorobenzene, alcohol and acetone. 52 Parts of a pure, strongly coloured, orange-yellow pigment are obtained which is distinguished by excellent fastness to light, migration and solvents.

The 3,3,4,5,6,7-hexachloro-isoindole-1-one is produced by heating 1 mol of tetrachlorophthalimide with 1.2 mol of phosphorus pentachloride in chlorobenzene for 1 hour at 125°. First the unchanged tetrachlorophthalimide is filtered off hot whereupon, on cooling, the new hexachloro-indolone crystallises out. After recrystallising from benzene, it melts at 205–206° on decomposition.

Calculated for $C_{12}HONCl_4$ N 4.11
Found N 4.27

Yellow pigments having similar good properties are obtained if in this example, the 1,4-diaminobenzene is replaced by 7.9 parts of 1,4-diaminonaphthalene or by 10.6 parts of 4,4'-diaminoazobenzene.

A redder pigment having the same good properties is obtained if in this example the 1,4-diaminobenzene is replaced by 12.75 parts of 3,3'-dichlorobenzidine. More greenish pigments having very good fastness properties are also obtained if in this example, the 1,4-diaminobenzene is replaced by 5.4 parts of 1,3-diaminobenzene, 6.1 parts of 1-methyl-2,6-

diaminobenzene, 12.3 parts of 2,7-diaminodibenzothioephene-1,1-dioxide or 11.5 parts of 4,4'-diamino-3-nitrodiphenyl.

EXAMPLE 2

A cold solution of 1.84 parts of 4,4'-diaminodiphenyl in 200 parts of benzene is added to a solution of 6.8 parts of 3,3,4,5,6,7-hexachloro-isoindole-1-one in 200 parts of benzene. An orange precipitate forms immediately. The whole is stirred for 1 hour at room temperature, the pigment is filtered off and washed with benzene, acetone and water. The pure orange pigment can be recrystallised from a great deal of nitrobenzene and melts at over 360°. It has excellent fastness to light as well as very good fastness to migration, solvents, cross-lacquering, oil, acid and alkali.

EXAMPLE 3

6.6 Parts of 3,3'-dichloro-4,4'-diaminodiphenyl di-hydrochloride are heated for half an hour at 200° with 11.6 parts of 3-imino-4,5,6,7-tetrachloro-isoindole-1-one in 200 parts of nitrobenzene. The yellow-brown pigment formed is filtered off hot from the nitrobenzene, washed with alcohol, water and acetone. It has good fastness to migration and light. The 3-imino-4,5,6,7-tetrachloro-isoindole-1-one is produced by heating 1 mol of tetrachlorophthalic acid anhydride and 4 mol of urea in the presence of 1/100th mol of ammonium molybdate in nitrobenzene at 140°. The not quite pure reaction product melts on decomposition around 305°; it provides a very valuable but not quite pure pigment.

EXAMPLE 4

A solution of 6.8 parts of 3,3,4,5,6,7-hexachloro-isoindole-1-one in 100 parts of xylene is refluxed for 2 hours with a solution of 2.44 parts of 4,4'-diamino-3,3'-dimethoxydiphenyl in 100 parts of xylene. The pigment is filtered off and washed with alcohol. It is a yellowish-red pigment which has very good fastness properties. If this pigment is again heated for 1 hour at 200° in 100 parts of nitrobenzene, it becomes bluish red and its already very good fastness to light is more noticeably improved.

EXAMPLE 5

A solution of 10.6 parts of 4,4'-diamino-3,3'-dimethyldiphenyl in 150 parts of *o*-dichlorobenzene and a solution of 35 parts of 3,3,4,5,6,7-hexachloro-isoindole-1-one in 500 parts of *o*-dichlorobenzene are heated for 3 hours at 160–170° while stirring. The precipitate is filtered off hot, washed with alcohol and acetone and, after drying, 34 parts of a pure red pigment are obtained which has very good fastness to oil, acid, alkali, solvents, migration and light.

Red pigments having the same good properties are also obtained if in this example, 7.4 parts of 1,5-diaminonaphthalene or 10.5 parts of 4,4'-diaminostilbene are used instead of 4,4'-diamino-3,3'-dimethyldiphenyl.

EXAMPLE 6

14.2 Parts of 3-imino-4,5,6,7-tetrachloro-isindole-1-one and 4.6 parts of 4,4'-diaminodiphenyl are refluxed for 2 hours in 300 parts of glacial acetic acid. A pure orange pigment is formed which is filtered off and purified by washing with glacial acetic acid, alcohol and acetone. The pigment is identical to that of example 2.

10 The 3-imino-4,5,6,7-tetrachloro-isindole-1-one used above is obtained by reacting 34 parts of 3,3,4,5,6,7-hexachloro-isindole-1-one with 60 parts of aqueous ammonia. It is a pale yellowish powder which melts at 320° on decomposition. The same pure orange pigment is obtained if, in this example, 18 parts of 3-phenylimino-4,5,6,7-tetrachloro-isindole-1-one (M.P. 264—265°) are reacted with 4.6 parts of 4,4'-diaminodiphenyl instead of 3-imino-4,5,6,7-tetrachloro-isindole-1-one. 3-Phenylimino - 4,5,6,7 - tetrachloro-isindole-1-one is obtained by reacting 34 parts of 3,3,4,5,6,7-hexachloro-isindole-1-one with 40 parts of aniline in benzene at 70°.

25 If in this example, the 3-imino-4,5,6,7-tetrachloro-isindole-1-one is replaced by 18 parts of 3-(2'-pyridylimino)-4,5,6,7-tetrachloro-isindole-1-one (M.P. 264—266°, produced from 3,3,4,5,6,7-hexachloro-isindole-1-one and 2-amino pyridine in *o*-dichlorobenzene), then the same orange pigment is obtained.

EXAMPLE 7

35 6.6 Parts of 1,3,3,4,5,6,7-heptachloro-isindolenine are dissolved in 80 parts of dioxan and the solution is added to a solution of 2 parts of 4,4'-diaminodiphenyl dihydrochloride in 300 parts of water. The suspension is heated to 80°, the orange pigment is filtered off and washed with alcohol and acetone. It can be recrystallised from nitrobenzene and is then identical to the pigment produced according to example 2.

EXAMPLE 8

45 6.8 Parts of 3,3,4,5,6,7-hexachloro-isindole-1-one are dissolved in 100 parts of acetone and the solution is added to 3.7 parts of 4,4'-diamino - stilbene - 2,2'-disulphonic acid dissolved in the form of the sodium salt in 200 parts of water. The addition is made at room temperature. The orange precipitate formed is filtered off, washed with water and dried. The dyestuff dissolves with a yellow colour in aqueous alkalis and dyes paper in orange-yellow shades.

50 If in this example, the 4,4'-diaminostilbene-2,2'-disulphonic acid is replaced by 3 parts of 3,3'-dicarboxy - 4,4'-diaminoazobenzene, a brown, water soluble dyestuff is obtained.

EXAMPLE 9

60 3.4 Parts of 3,3,4,5,6,7-hexachloro-isindole-1-one and 1 part of hydrazine hydrate are heated to 100° for 5 minutes in 100 parts of chlorobenzene. The greenish-yellow pigment which precipitates is filtered off hot and

washed with acetone and water. It is distinguished by its purity and its excellent fastness to migration and light.

EXAMPLE 10

70 1.58 Parts of pyridine are added to 6.8 parts of 3,3,4,5,6,7-hexachloro-isindole-1-one in 100 parts of dioxan. A white precipitate immediately forms. A solution of 1.84 parts of 4,4'-diaminodiphenyl in 50 parts of dioxan is added whereupon the suspension turns first yellow and then orange. After heating for 2 hours at 90—95°, the orange pigment is filtered off and washed with alcohol and water. It has excellent fastness to light.

EXAMPLE 11

80 The solution of a mixture of 1.84 parts of 4,4'-diaminodiphenyl and 2.53 parts of 3,3'-dichlorobenzidine in 100 parts of *o*-dichlorobenzene is added to a solution of 13.2 parts of 3,3,4,5,6,7-hexachloro-isindole-1-one in 200 parts of *o*-dichlorobenzene. The orange suspension formed is heated for an hour at 170°, the precipitate is filtered off and washed with acetone. The pigment thus obtained is more yellow but has the same good fastness properties as that of example 2.

EXAMPLE 12

95 2.38 Parts of 1,5-diamino anthraquinone are dissolved in 100 parts of hot nitrobenzene and condensed for 2 hours at 140° with 6.6 parts of 3,3,4,5,6,7-hexachloro-isindole-1-one dissolved in 100 parts of nitrobenzene. The pigment which has been filtered off is stirred again for 1 hour at 200° in 100 parts of nitrobenzene, filtered off hot and washed with alcohol and acetone. The pigment so obtained is blue-black and is distinguished by its good fastness properties and its colour strength.

EXAMPLE 13

105 13 Parts of 3,3,4,5,6,7-hexachloro-isindole-1-one are heated with 2.2 parts of 2,6-diaminopyridine in 200 parts of *o*-dichlorobenzene for 2 hours at 170°. The yellow pigment is filtered off hot and washed with alcohol. It has good fastness to acid, alkali, oil and light.

EXAMPLE 14

110 10 Parts of 3,3,4,5,6,7-hexachloro-isindole-1-one are dissolved in 150 parts of *o*-dichlorobenzene and the solution is mixed with a solution of 2 parts of 2,4,4'-triiminodiphenyl in 100 parts of *o*-dichlorobenzene. The whole is heated for 2 hours at 150°. A reddish-yellow pigment which has good fastness properties is obtained.

EXAMPLE 15

120 142.5 Parts of tetrachlorophthalimide and 110 parts of phosphorus pentachloride in 500 parts of chlorobenzene are heated for 1 hour at 125°. A solution of 48 parts of 4,4'-diaminodiphenyl in 300 parts of chlorobenzene is then added and the whole is stirred for 2 hours at 125°. 300 Parts of ethanol are added to the suspension, the pigment is filtered off, washed with alcohol, acetone, dilute ammonia solution and water and dried. This

crude pigment is again purified by boiling out with nitrobenzene and filtering hot. The pigment is orange and has good fastness to migration and light.

EXAMPLE 16

- 2.5 Parts of a mixture of 3-imino-4,5,7- and 4,6,7-trichloroisindole-1-one are heated with 0.5 parts of 1,4-diaminobenzene in 50 parts of naphthalene for 1 hour at 220°. (The 3-imino-4,5,7- and 4,6,7-trichloroisindole-1-one is produced by heating 10 parts of 3,4,6-trichlorophthalic acid anhydride, 20 parts of urea and 0.2 parts of ammonium molybdate in 100 parts of nitrobenzene at 140°, filtering off the precipitate and purifying by extraction with water at 60°). The melt is then diluted with 100 parts of *o*-dichlorobenzene, filtered and the residue is washed with alcohol and acetone. A yellow pigment which has good fastness to light is obtained in this way.

EXAMPLE 17

- 17.2 Parts of 3,3,4,5,6,7-hexachloroisindole-1-one are dissolved in 250 parts of benzene. This solution is mixed with a solution of 4.6 parts of 4,4'-diaminodiphenyl in 200 parts of benzene and 8 parts of pyridine and the whole is refluxed for 1 hour. A pure orange pigment is formed which is filtered off under suction and washed with alcohol and water.

Also 10.1 parts of triethylamine can be used in this example instead of pyridine.

EXAMPLE 18

- 4.41 Parts of 3,3-bis-morpholino-4,5,6,7-tetrachloroisindole-1-one (M.P. 211—212°, produced by reacting 3,3,4,5,6,7-hexachloroisindole-1-one with morpholine in benzene), are heated for 5 minutes at 100° with 0.92 parts of 4,4'-diaminodiphenyl in 50 parts of glacial acetic acid. After filtering and washing the residue with glacial acetic acid and water, an orange pigment is isolated which has excellent fastness properties.

- An orange pigment having similar good properties is obtained if in this example, the 3,3-bis-morpholino-4,5,6,7-tetrachloroisindole-1-one is replaced by 5.25 parts of 3,3-bis-dibutylamino-4,5,6,7-tetrachloroisindole-1-one. This latter compound is produced by reacting 3,3,4,5,6,7-hexachloroisindole-1-one with di-*n*-butylamine in benzene.

EXAMPLE 19

- 3.31 Parts of 3,3-dimethoxy-4,5,6,7-tetrachloroisindole-1-one (M.P. 165—175° on decomposition, produced from 3,3,4,5,6,7-hexachloroisindole-1-one and sodium methylate in methyl alcohol), are heated for 2 hours at 150° with 0.54 parts of 1,4-diaminobenzene in 50 parts of *o*-dichlorobenzene. A yellow pigment is obtained which has very good fastness properties.

EXAMPLE 20

- 3.59 Parts of 3,3-diethoxy-4,5,6,7-tetrachloroisindole-1-one (M.P. 175—180° on

decomposition, produced from 3,3,4,5,6,7-hexachloroisindole-1-one and sodium ethylate in ethyl alcohol), are boiled for 30 minutes with 0.92 parts of 4,4'-diaminodiphenyl in 50 parts of glacial acetic acid. An orange pigment is obtained which has excellent fastness properties.

EXAMPLE 21

2.96 Parts of 2,5-bis-(4'-aminophenyl)-oxadiazole, and 6.8 parts of 3,3,4,5,6,7-hexachloroisindole-1-one in 100 parts of *o*-dichlorobenzene are heated for 2 hours in a vacuum at 120° under 90 mm Hg pressure. After filtering off and washing the residue with alcohol, acetone and water, a pure yellow pigment is obtained which has excellent fastness to light and migration.

If, instead of 2,5-bis-(4'-aminophenyl)-oxadiazole, 3.20 parts of 2,5-bis-(3'-methyl-4'-aminophenyl)-oxadiazole are used, then a more reddish pigment which has similar good properties is obtained.

Pigments having still more red shades are obtained if in this example the 2,5-bis-(4'-aminophenyl)-oxadiazole is replaced by 3.12 parts of 2,5-bis-(4'-aminophenyl)-thiadiazole or by 3.8 parts of 2,5-bis-(3'-chloro-4'-aminophenyl)-thiadiazole. The parts of the oxadiazoles and thiadiazoles given above are intentionally somewhat in excess of the stoichiometric amounts that would give similar results.

EXAMPLE 22

10 Parts of the pigment obtained according to example 1 from 1,4-diaminobenzene, 10 parts of hydrate of alumina, 30 parts of linseed oil varnish of medium viscosity and 0.2 parts of cobalt linoleate are mixed and rubbed in a three-roll mill. A graphic colour is obtained which produces reddish-yellow prints of particular purity and colour strength and, in addition, has excellent fastness to solvents, acid, alkali and light.

EXAMPLE 23

10 Parts of the pigment obtained according to example 2, 10 parts of titanium dioxide, 35 parts of a 60% solution of a modified urea allyl resin in xylene/butanol 1:1, 10 parts of turpentine oil and 5 parts of xylene are milled for 45 hours in a ball mill. After pouring onto aluminium sheets and annealing for one hour at 120° a pure orange dyeing is obtained which has excellent fastness properties, in particular very good fastness to cross lacquering.

EXAMPLE 24

67 Parts of polyvinyl chloride, 33 parts of dioctyl phthalate, 2 parts of dibutyl tin dilaurate, 1 part of the pigment obtained according to example 4 and 2.5 parts of titanium dioxide are mixed for 15 minutes at 150° on a roller frame and worked up into a thin foil. The red dyeing so obtained is distinguished by its good colour strength and its excellent fastness to heat, migration and light.

WHAT WE CLAIM IS:—

1. Process for the production of coloured

condensation products characterised by condensing polyprimary amino compounds of the homo- or hetero-cyclic aromatic series or hydrazine with a molecular equivalent for every primary amino group thereof of the same or different isoindole-1-ones containing at least 3 halogen atoms in the benzo radical, which isoindole-1-ones contain in the 3-position an imino group which can be organically substituted, a doubly bound sulphur atom, two halogen atoms, two secondary amino groups or two ether groups.

2. Process according to claim 1 characterised by the use of diprimary amino compounds of the homo- or hetero-cyclic aromatic series.

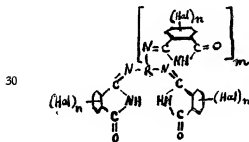
3. Process according to claim 1 characterised by the use of hydrazine as a diprimary amino compound.

4. Process according to claim 1 characterised by the use of two different polyprimary amino compounds.

5. Process according to claim 1 characterised by the use of such isoindole-1-ones which contain in the 3-position two alkoxy groups.

6. Process according to claims 1 and 4 characterised by the use of 3,3,4,5,6,7-hexachloro-isoindole-1-one.

7. A coloured condensation product of the formula:

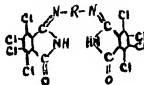


wherein Hal represents a halogen atom,
R represents a direct linkage when m is

nought or the radical of an organic compound of the homo- or hetero-cyclic aromatic series, the nitrogen atoms are directly bound to the aromatic nucleus of the radical,

n represents the number 3 or 4 and m is one of the numerals 0 and 1.

8. A coloured condensation product of the formula:



wherein $=N-R-N=$ represents the radical of a diprimary amino benzene, diphenyl, stilbene or naphthalene compound.

9. Manufacture of coloured condensation products substantially as described with reference to any of the foregoing Examples 1 to 21.

10. Coloured condensation products as defined in claim 1 whenever prepared or produced by the processes of manufacture hereinbefore particularly described.

11. A coloured condensation product as specified in any of the foregoing Examples 1 to 21 including a condensation product resulting from an alternative reactant specified.

12. Process for the dyeing and pigmentation of cellulose, plastics and lacquers characterised by the use of coloured condensation products according to claim 6 and substantially as described in Examples 22 to 24.

13. Cellulose, plastics, and lacquers dyed or pigmented by the use of coloured condensation products defined in claim 6.

For the Applicants,
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